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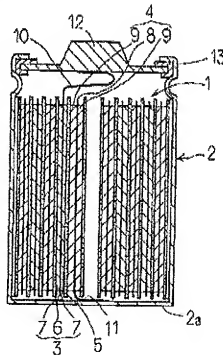
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(54) LITHIUM ION SECONDARY BATTERY AND ITS MANUFACTURE

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a lithium ion secondary battery capable of increasing high rate discharge property of the battery and the number of life cycles with the wettability of electrolyte inside a pole material layer enhanced sufficiently.

SOLUTION: Positive electrode material slurry containing lithium contained double oxide and organic solvent is applied to a positive electrode collector 6, then dried, and pressed to form a positive electrode plate 3. A negative electrode material slurry containing carbon material and organic solvent is applied onto a negative electrode collector 8 and then dried and pressed to form a negative electrode plate 4. The positive electrode plate 3 and the negative electrode plate 4 are laminated via a separator 5 to form a pole plate group 1, which is impregnated with a nonaqueous electrolyte. Amphoteric surface active agent, such as carboxylic acid salt type surface active agent, is added to at least one of the positive electrode material slurry, the negative electrode material slurry and the nonaqueous electrolyte.



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CLAIMS

[Claim(s)]

[Claim 1]In a rechargeable lithium-ion battery which it comes to laminate via a nonaqueous electrolyte layer in which a positive electrode material layer which uses a lithium content multiple oxide as the main ingredients, and a negative electrode material layer which makes a lithium ion occlusion and uses a substance to emit as the main ingredients contain lithium salt, at least one of said positive electrode material layer, said negative electrode material layer, and said the nonaqueous electrolyte layers -- an ampholytic surface active agent -- ***** -- a rechargeable lithium-ion battery characterized by its.

[Claim 2]The rechargeable lithium-ion battery according to claim 1, wherein said ampholytic surface active agent is a carboxylate type surface-active agent.

[Claim 3]In a rechargeable lithium-ion battery which it comes to laminate via a nonaqueous electrolyte layer in which a positive electrode material layer which uses a lithium content multiple oxide as the main ingredients, and a negative electrode material layer which makes a lithium ion occlusion and uses a substance to emit as the main ingredients contain lithium salt, A rechargeable lithium-ion battery, wherein 0.01 to 0.2% of the weight of a carboxylate type surface-active agent is contained in said positive electrode material layer to said lithium content multiple oxide.

[Claim 4]In a rechargeable lithium-ion battery which it comes to laminate via a nonaqueous electrolyte layer in which a positive electrode material layer which uses a lithium content multiple oxide as the main ingredients, and a negative electrode material layer which makes a lithium ion occlusion and uses a carbon material to emit as the main ingredients contain lithium salt, A rechargeable lithium-ion battery, wherein 0.01 to 0.2% of the weight of a carboxylate type surface-active agent is contained in said negative electrode material layer to said carbon material.

[Claim 5]In a rechargeable lithium-ion battery which it comes to laminate via a nonaqueous electrolyte layer in which a positive electrode material layer which uses a lithium content multiple oxide as the main ingredients, and a negative electrode material layer which makes a lithium ion occlusion and uses a carbon material to emit as the main ingredients contain lithium salt, A rechargeable lithium-ion battery, wherein 0.01 to 0.2% of the weight of a carboxylate type surface-

active agent is contained in said nonaqueous electrolyte layer to said lithium salt.

[Claim 6]After applying positive electrode material slurry containing a lithium content multiple oxide and an organic solvent on a positive pole collector, Make an anode board provided with an anode agent layer through a drying process and press working of sheet metal, and a lithium ion Occlusion, After applying a negative electrode material slurry containing a substance to emit and an organic solvent on a negative pole collector, Make a negative electrode plate provided with a negative electrode material layer through a drying process and press working of sheet metal, and laminate said anode board and said negative electrode plate via a separator, and a group of electrode is made, A manufacturing method of a rechargeable lithium-ion battery adding an ampholytic surface active agent in a method of impregnating said group of electrode with nonaqueous electrolyte, and manufacturing a rechargeable lithium-ion battery, to at least one of said positive electrode material slurry, a negative electrode material slurry, and the nonaqueous electrolyte.

[Claim 7]A manufacturing method of the rechargeable lithium-ion battery according to claim 6 using a carboxylate type ampholytic surface active agent as said ampholytic surface active agent.

[Claim 8]In a manufacturing method of an anode board for rechargeable lithium-ion batteries which makes an anode board provided with an anode agent layer through a drying process and press working of sheet metal after applying positive electrode material slurry containing a lithium content multiple oxide, an organic solvent, and electric conduction powder on a positive pole collector, A manufacturing method of an anode board for rechargeable lithium-ion batteries adding an ampholytic surface active agent to said positive electrode material slurry.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to a rechargeable lithium-ion battery and a manufacturing method for the same.

[0002]

[Description of the Prior Art]Generally, the positive electrode material layer which consists of a lithium content multiple oxide, and the negative electrode material layer which consists a lithium ion of occlusion and a carbon material to emit are laminated via a nonaqueous electrolyte layer, and the rechargeable lithium-ion battery is constituted. This rechargeable lithium-ion battery is manufactured as follows. First, the organic solvent which consists of the positive electrode material and binder which consist of a lithium content multiple oxide, N-methyl-2-pyrrolidone (NMP), etc. is kneaded, and positive electrode material slurry is made. And after applying this positive electrode material slurry on a positive pole collector, the anode board provided with the anode agent layer through a drying process and press working of sheet metal is made. The organic solvent which consists a lithium ion of the negative electrode material and binder which consist of occlusion and a carbon material to emit, NMP, etc. is kneaded, and a negative electrode material slurry is made. And after applying this negative electrode material slurry on a negative pole collector, the negative electrode plate provided with the negative electrode material layer through a drying process and press working of sheet metal is made. Next, an anode board and a negative electrode plate are laminated via a separator, a group of electrode is made, and this group of electrode is put into a battery case. And this group of electrode is impregnated with the nonaqueous electrolyte which dissolved lithium salt, such as lithium hexafluoride phosphorate, in organic solvents, such as carbonic ester, and a cell is completed. The rechargeable lithium-ion battery has high energy density.

Since self-discharge is small, it is widely used as a portable power supply of a miniaturization and the electronic equipment by which the weight saving was carried out.

However, in this rechargeable lithium-ion battery, when making positive electrode material slurry

and a negative electrode material slurry, there is a problem that the dispersibility to the inside of the organic solvent of a positive electrode material and a negative electrode material is bad. There is a problem that the wettability over the positive electrode material layer and negative electrode material layer of nonaqueous electrolyte is bad. A wettable fall is remarkable when carrying out compression molding of the polar plate to a thickness direction with a roll press machine especially at the time of polar-plate manufacture. Then, as shown in JP,H9-306501,A for a wettable improvement, adding the cationic surface active agent which becomes either [at least] a positive electrode material layer or a negative electrode material layer from oleic amide was proposed. If oleic amide is added, oleic amide will be eluted to the inside of the opening inside a polar plate, and a plate surface, and the wettability of the whole polar plate will become high.

[0003]

[Problem(s) to be Solved by the Invention]However, even if it added oleic amide in this way to either [at least] the positive electrode material layer or the negative electrode material layer, there is a limit in improving the wettability of the electrolysis solution inside a polar plate, and the high rate discharging characteristic and the number of life cycles of the cell were not able to be raised substantially. Even if especially positive electrode material slurry put in a cationic surface active agent like oleic amide in order to show alkalinity, it was not able to improve dispersibility to the inside of the organic solvent of a positive electrode material.

[0004]The purpose of this invention is to provide a rechargeable lithium-ion battery which can fully improve the wettability of the electrolysis solution inside an electrode material layer, and can raise the high rate discharging characteristic and the number of life cycles of a cell, and a manufacturing method for the same.

[0005]Other purposes of this invention are to provide the manufacturing method of the rechargeable lithium-ion battery to which dispersibility to the inside of the organic solvent of the positive electrode material at the time of making positive electrode material slurry is made as for high ***** in addition to the above-mentioned purpose.

[0006]

[Means for Solving the Problem]A positive electrode material layer which uses a lithium content multiple oxide as the main ingredients, and a negative electrode material layer which makes a lithium ion occlusion and uses a substance to emit as the main ingredients make this invention an object of improvement of a rechargeable lithium-ion battery which it comes to laminate via a nonaqueous electrolyte layer. And at least one of a positive electrode material layer, a negative electrode material layer, and the nonaqueous electrolyte layers is made to contain an ampholytic surface active agent in this invention. An ampholytic surface active agent here is a surface-active agent provided with character of both an anionic surface active agent and a cationic surface active agent. The ampholytic surface active agent can improve the wettability of an electrolysis solution inside a polar plate compared with cationic surface active agents, such as conventional oleic amide. Therefore, a high rate discharging characteristic and the number of life cycles of a rechargeable lithium-ion battery are raised rather than a case where a cationic surface active agent is used.

[0007]As an ampholytic surface active agent, there are a carboxylate type, a sulfuric-ester-salt type, a sulfonate type, an phosphate type, etc. Many especially carboxylate type ampholytic surface active agents are marketed, and there is an advantage that composition can be easily done compared with other things. As a carboxylate type ampholytic surface active agent, an amine type whose cation portion is amine salt, or a betaine type whose cation portion is quarternary ammonium salt can be used.

[0008]As a lithium content multiple oxide to be used, lithium, a transition metal, and an included oxide can be used as a positive electrode material. As a transition metal, a kind is [choose / out of Ti, V Cr, Mn, Fe, Co, nickel, Mo, W, and Cu] employable in it being few.

[0009]As a substance used as a negative electrode material, scaly natural graphite, mesophase pitch system black lead, a massive artificial graphite, etc. can be used as black lead. As amorphous carbon material, meso carbon micro beads, a furfuryl-alcohol-resin baking body, etc. can be used.

[0010]What dissolved an electrolyte which consists of lithium salt in an organic solvent as nonaqueous electrolyte is used. As an organic solvent, propylene carbonate, ethylene carbonate, 1,2-JIMEKISHI ethane, 1,2-JIEKISHI ethane, 2-methyltetrahydrofuran, A thing chosen from diethyl carbonate, gamma-butyllactone, a tetrahydrofuran, diethylether, sulfolane, acetonitrile, etc. or a thing which mixed these can be used. As lithium salt, LiClO_4 , LiPF_6 , LiBF_4 , LiCl , LiBr , $\text{CH}_3\text{SO}_3\text{Li}$, $\text{Li}(\text{CF}_3\text{SO}_2)_2\text{N}$, $\text{Li}(\text{C}_2\text{F}_5\text{SO}_2)_2\text{N}$, LiAsF_6 , etc. can be used.

[0011]When making a positive electrode material layer contain a carboxylate type ampholytic surface active agent, it is preferred to make it contain 0.01 to 0.2% of the weight to a lithium content multiple oxide of a positive electrode material layer. If less than 0.01 % of the weight, the wettability of an electrolysis solution inside an anode board becomes insufficient, and capacity cannot be raised. When it exceeds 0.2 % of the weight, there is a problem to which a fill ration of a positive electrode material falls to, and capacity falls.

[0012]When making a negative electrode material layer contain a carboxylate type ampholytic surface active agent, it is preferred to make it contain 0.01 to 0.2% of the weight to a carbon material of a negative electrode material layer. If less than 0.01 % of the weight, the wettability of an electrolysis solution inside a negative electrode plate becomes insufficient, and capacity cannot be raised. When it exceeds 0.2 % of the weight, there is a problem to which a fill ration of a positive electrode material falls to, and capacity falls.

[0013]When making a nonaqueous electrolyte layer contain a carboxylate type ampholytic surface active agent, it is preferred to make it contain 0.01 to 0.2% of the weight to lithium salt of a nonaqueous electrolyte layer. If less than 0.01 % of the weight, the wettability of an electrolysis solution an anode board and inside a negative electrode plate becomes insufficient, and capacity cannot be raised. When it exceeds 0.2 % of the weight, there is a problem to which the lithium conductivity of a nonaqueous electrolyte layer falls to, and capacity falls.

[0014]In order to manufacture a rechargeable lithium-ion battery of this invention, after applying positive electrode material slurry containing a lithium content multiple oxide and an organic solvent

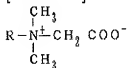
on a positive pole collector first, an anode board provided with an anode agent layer through a drying process and press working of sheet metal is made. After applying a negative electrode material slurry which contains occlusion, a carbon material to emit, and an organic solvent for a lithium ion on a negative pole collector, a negative electrode plate provided with a negative electrode material layer through a drying process and press working of sheet metal is made. Next, an anode board and a negative electrode plate are laminated via a separator, a group of electrode is made, this group of electrode is impregnated with nonaqueous electrolyte, and a rechargeable lithium-ion battery is manufactured. And an ampholytic surface active agent is added to at least one of positive electrode material slurry, a negative electrode material slurry, and the nonaqueous electrolyte. Thus, if a rechargeable lithium-ion battery is manufactured, a rechargeable lithium-ion battery which has the wettability of an electrolysis solution inside a polar plate improved can be manufactured easily. In order that especially positive electrode material slurry may show alkalinity, when an ampholytic surface active agent is added to positive electrode material slurry, it has dispersibility to inside of an organic solvent of a positive electrode material improved. Since positive electrode material slurry contains electric conduction powder, if dispersibility is improved in this way, an electric conduction network of electric conduction powder will be uniformly formed into a positive electrode material layer. Therefore, a reaction in a positive electrode material layer equalizes, and movement of ion becomes easy. As a result, charge transfer resistance becomes low and the high rate discharging characteristic of a cell improves.

[0015]

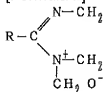
[Embodiment of the Invention](Examination 1) Drawing 1 is an end elevation of each rechargeable lithium-ion battery used for the examination 1. As shown in this figure, this rechargeable lithium-ion battery has the structure where the winding type group of electrode 1 was stored in the battery can 2. And the winding type group of electrode 1 has the structure wound so that the anode board 3 and the negative electrode plate 4 might laminate via the electrolyte layer (separator) 5. In this example, the rechargeable lithium-ion battery was manufactured as follows. The anode board 3 was manufactured first. First, the positive electrode material which consists of cobalt acid lithium (Li_xCoO_2) with a mean particle diameter of 10 micrometers, The electric conduction auxiliary agent which consists of the carbon powder end of 3-micrometer mean particle diameter, and the ampholytic surface active agent of the amount of each (weight to cobalt acid lithium of a positive electrode material) shown in the binder which consists of polyvinylidene fluorides, and Table 1 were distributed to the solvent which consists of N-methyl-2-pyrrolidone (NMP), and the anode slurry was made. Here in the ampholytic surface active agent shown in Table 1 REBON 2000 (fluid) and NSA-2000 (fluid), Mitsuhiro -- transformation -- it is an incorporated company betaine type amphoteric surfactant -- REBON 101-H (fluid) and REBON 105 (fluid) -- Mitsuhiro -- transformation -- it is an incorporated company isoDAZORIN type ampholytic surface active agent. A fundamental structure of a betaine type amphoteric surfactant is shown in following ** 1, and a fundamental structure of an isoDAZORIN type ampholytic surface active agent is shown in following ** 2.

[0016]

[Formula 1]



[Formula 2]



Next, after applying the anode slurry to thickness uniform to both sides of the positive pole collector 6 which consists of 20-micrometer-thick aluminium foil, it dried and NMP was removed, it rolled with the roll press machine, the positive electrode material layer 7 was formed, and the anode board (480 mm in length, 54 mm in width, and 174 micrometers in thickness) 3 was made.

[0017]Next, the negative electrode plate 4 was manufactured. First, the negative electrode material which consists of a carbon material of black lead with a mean particle diameter of 20 micrometers, and the binder which consists of polyvinylidene fluorides were distributed to the solvent which consists of N-methyl-2-pyrrolidone (NMP), and the negative-electrode slurry was made. Next, after applying the negative-electrode slurry to thickness uniform to both sides of the negative pole collector 8 which consists of 10-micrometer-thick copper foil, it dried and NMP was removed, it rolled with the roll press machine, the negative electrode material layer 9 was formed, and the negative electrode plate (500 mm in length, 56 mm in width, and 174 micrometers in thickness) 4 was made.

[0018]Next, the anode board 3 and the negative electrode plate 4 were wound via the band-like separator 5 which consists of 24-micrometer-thick polyethylene fine porous membrane, and the group of electrode 1 was made. The band-like separator 5 is constituted by separator part of a couple. And a separator part of a couple was arranged and wound around a diameter direction lateral part and a diameter direction inner part of winding which adjoin the battery can 2 to both sides of the negative electrode plate 4 so that a separator might be arranged. Next, after having arranged the group of electrode 1 in the battery can 2 of a cylindrical shape which consists of nickel plating iron, the nickel tab terminal 11 currently beforehand welded to the negative pole collector 8 was welded to the pars basilaris ossis occipitalis 2a of the battery can 2. Next, 5 ml of organic electrolysis liquid (nonaqueous electrolyte) which dissolved lithium salt which consists of LiPF_6 by concentration of 1 mol/l. in a solvent which mixed propylene carbonate and dimethyl carbonate by the volume ratio 1:1 was poured in into the battery can 2. Next, the aluminum tab terminal 10 currently beforehand welded to the positive pole collector 6 was welded to the battery lid 12 provided with a pressure switch. And after having arranged the battery lid 12 in the upper part of the battery can 2 via the gasket 13 which consists of insulating polypropylene, inside of the

battery can 2 was sealed for this in total, and each uncharged rechargeable lithium-ion battery of a cylindrical shape of 18 mm in diameter and 65 mm[in height] , was made.

[0019]Next, after charging each uncharged rechargeable lithium-ion battery by the programmed voltage 4.2V and 1400 mA of limiting current in 25 ** for 2.5 hours, service capacity of each cell at the time of discharging to 2.5V at 1400 mA was calculated. Above-mentioned charge and discharge were repeated, service capacity after 500 cycles was measured, and a rate (charge-discharge cycle characteristic) over service capacity at the time of first discharge of this service capacity was computed. Table 1 shows the result. A result of what added oleic amide used as a comparative example a thing (additive-free) which does not add an additive agent, and conventionally is also collectively shown in Table 1.

[0020]

[Table 1]

添 加 剤	添加量							
	0.005wt%		0.01wt%		0.2wt%		0.25wt%	
	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)
レボソ2000	1480	70	1550	81	1560	82	1500	69
NSA-2000	1490	72	1580	82	1570	83	1490	68
レボソ101-H	1458	69	1560	81	1570	82	1510	69
レボソ105	1497	68	1590	79	1575	81	1505	70
オレイン酸アミド	1510	72	1515	75	1513	73	1520	76
無添加	1487	71	1487	71	1487	71	1487	71

Compared with what added oleic amide used additive-free and conventionally, with a rechargeable lithium-ion battery which made a positive electrode material layer contain an ampholytic surface active agent 0.01 to 0.2% of the weight, service capacity is raised and, moreover, a charge-discharge cycle characteristic is understood that it can improve from Table 1.

[0021](Examination 2) Adding an ampholytic surface active agent of the amount of each (weight to a carbon material of a negative electrode material) shown in Table 2 to a negative electrode material slurry instead of positive electrode material slurry next, others created a cell used for the examination 1, and same cell, and asked for service capacity and a charge-discharge cycle characteristic of each cell by the same test condition as the examination 1. Table 2 shows the result. A result of what added oleic amide used as a comparative example a thing (additive-free) which does not add an additive agent, and conventionally is also collectively shown also in Table 2.

[0022]

[Table 2]

添 加 剤	添加量							
	0.005wt%		0.01wt%		0.2wt		0.25wt	
	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)
レボソ2000	1480	72	1550	85	1560	87	1500	71
NSA-2000	1490	71	1580	86	1570	85	1490	70
レボソ101-H	1458	71	1560	88	1570	88	1510	72
レボソ105	1497	72	1590	89	1575	89	1505	69
オレイン酸アミド	1521	78	1531	80	1532	81	1520	74
無添加	1487	71	1487	71	1487	71	1487	71

Compared with what added the oleic amide used additive-free and conventionally, with the rechargeable lithium-ion battery which made the negative electrode material layer contain an ampholytic surface active agent 0.01 to 0.2% of the weight, service capacity is raised and, moreover, a charge-discharge cycle characteristic is understood that it can improve from Table 2. [0023](Examination 3) Adding the ampholytic surface active agent of the amount of each (weight to lithium salt of nonaqueous electrolyte) shown in nonaqueous electrolyte instead of positive electrode material slurry below in Table 3, others created the cell used for the examination 1, and the same cell, and asked for the service capacity and the charge-discharge cycle characteristic of each cell by the same test condition as the examination 1. Table 3 shows the result. The result of what added the oleic amide used as a comparative example the thing (additive-free) which does not add an additive agent, and conventionally is also collectively shown also in Table 3.

[0024]

[Table 3]

添 加 剤	添加量							
	0.005wt%		0.01wt%		0.2wt		0.25wt	
	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)	放電容量 (mAh)	サイクル 特性 (%)
レボソ2000	1480	67	1550	78	1560	77	1500	68
NSA-2000	1490	68	1580	76	1570	76	1490	67
レボソ101-H	1458	70	1560	75	1570	78	1510	69
レボソ105	1497	69	1590	77	1575	75	1505	70
オレイン酸アミド	1450	65	1461	67	1465	68	1453	66
無添加	1487	71	1487	71	1487	71	1487	71

Compared with what added the oleic amide used additive-free and conventionally, with the rechargeable lithium-ion battery which made the negative electrode material layer contain an ampholytic surface active agent 0.01 to 0.2% of the weight, service capacity is raised and, moreover, a charge-discharge cycle characteristic is understood that it can improve from Table 3.

[0025]Tables 1-3 show a thing especially with a high effect, when an ampholytic surface active agent is added to a positive electrode material layer and a negative electrode material layer.

[0026]Although the ampholytic surface active agent was added in above-mentioned working

example, respectively to each part of the positive electrode material layer, the negative electrode material layer, and the nonaqueous electrolyte layer, What is necessary is just to add an ampholytic surface active agent to at least one of a positive electrode material layer, a negative electrode material layer, and the nonaqueous electrolyte layers, and, of course, an ampholytic surface active agent may be added in all a positive electrode material layer and a negative electrode material layer or positive electrode material layers, the negative electrode material layers, and nonaqueous electrolyte layers.

[0027]Although the battery can of the group of electrode and cylindrical shape which were wound was used in above-mentioned working example, the same effect can be acquired also in the cell using the battery can of the group of electrode which only laminated the tabular polar plate and constituted it, and multiple pillars (a triangular prism, a square pole, etc.).

[0028]At this example, although amorphous carbon material was used as a negative electrode material, of course, black lead may be used as a negative electrode material.

[0029]

[Effect of the Invention]Since at least one of a positive electrode material layer, a negative electrode material layer, and the nonaqueous electrolyte layers is made to contain an ampholytic surface active agent according to this invention, the wettability of the electrolysis solution inside a polar plate can be improved, and the high rate discharging characteristic and the number of life cycles of a rechargeable lithium-ion battery are raised. In order that especially positive electrode material slurry may show alkalinity, when an ampholytic surface active agent is added to positive electrode material slurry, it has the dispersibility to the inside of the organic solvent of a positive electrode material improved. Since positive electrode material slurry contains electric conduction powder, if dispersibility is improved in this way, the electric conduction network of electric conduction powder will be uniformly formed into a positive electrode material layer. Therefore, the reaction in a positive electrode material layer equalizes, and movement of ion becomes easy. As a result, charge transfer resistance becomes low and the high rate discharging characteristic of a cell improves.

[Translation done.]